

Adiabatic pumping through a quantum dot with coulomb interactions: A perturbation expansion in the tunnel coupling

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We present a diagrammatic real-time approach to adiabatic pumping of electrons through interacting quantum dots. Performing a systematic perturbation expansion in the tunnel-coupling strength, we compute the charge pumped through a single-level quantum dot per pumping cycle. The combination of Coulomb interaction and quantum fluctuations, accounted for in contributions of higher order in the tunnel coupling, modifies the pumping characteristics via an interaction-dependent renormalization of the quantum-dot level. The latter is even responsible for the *dominant* contribution to the pumped charge when pumping via time-dependent tunnel-coupling strengths.

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I. INTRODUCTION

In the absence of an applied bias voltage, a mesoscopic conductor can sustain a DC current component if two or more parameters of the device (for example gate voltages) are periodically modulated in time. Electron *pumping* is said to be *adiabatic* if the parameter variation is slow on the scale defined by the dwell time of the electrons. In this case the pumped charge depends on the size and the shape of the pumping cycle but not on its detailed time evolution, i.e. it is of geometric nature.¹ The idea of electron pumping dates back to a work of Thouless.² The first experiment on electron pumping in single electron devices was performed by Pothier *et al.*³ Since then much theoretical and experimental work has been devoted to electron pumping.^{1,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28,29,30,31}

For non-interacting systems the theory of adiabatic pumping, formulated by Brouwer,⁴ is based on the generalization³² of the scattering approach for quantum transport to time-dependent phenomena. This formulation has been applied to study several aspects of pumping in non-interacting systems, such as the study of noise and decoherence,^{7,10,11} the role of discrete symmetries,⁹ the possibility of spin pumping,^{12,13,14} and the effect of superconducting elements and Andreev reflection.^{15,16,17} A diagrammatic approach was used in Refs. 18,19,20 to calculate the pumped charge through a noninteracting system using random matrix theory in the limit of a large number of channels in the leads. Furthermore several works investigate pumping by surface acoustic waves both theoretically⁶ and experimentally.²⁴ Pumping in interacting systems has been studied much less so far. Quantum pumping through a Luttinger

liquid has been discussed.²⁹ In quantum dots pumping has been studied in the limit of weak interaction,^{25,26} in the Kondo regime,²⁷ as well as in the Coulomb-blockade regime.²⁸ In Refs. 25,26 the pumped charge through an open quantum dot is computed by means of the bosonization technique. Aono, in Ref. 27 uses the Keldysh Green's function formalism complemented by the assumption that the dot retarded Green's function takes the non interacting form (this holds true in the non-interacting limit as well as in the Kondo regime). The authors of Ref. 28 integrate numerically the master equation (in the Born-Markov approximation) for the reduced density matrix of a double-dot pump. Recently a general approach to pumping through interacting quantum dots has been put forward by relating the pumped charge to the instantaneous retarded Green's functions of the quantum dot.^{30,31}

In this paper we study adiabatic pumping through interacting quantum dots for temperatures much above the Kondo temperature but much below the level spacing in the dot. In this case a perturbative expansion in the tunnel coupling between the dot and the leads is justified. Moreover we can restrict ourselves to consider only one level in the dot with a strong local repulsion in the case of double occupancy. We aim at the understanding of the influence of Coulomb interaction on the pumping characteristics. In order to achieve this, we extend a diagrammatic real-time technique³³ that has been developed to describe non-equilibrium DC transport through an interacting quantum dot. As compared to the formalism in our recent work,³⁰ the perturbative approach presented here, although limited to weak tunnel-coupling strengths, is more transparent in identifying the physical origin of the various contributions to the pumped charge. In par-

ticular it is straightforward to relate the pumped current to the dynamics of the average charge of the dot.

In this work we calculate the leading- and next-to-leading-order contribution of the perturbation expansion in the tunnel coupling to the pumped charge per pumping cycle. We distinguish two cases: pumping by changing periodically either the gate voltage and one tunnel barrier or varying both left and right tunnel barrier. Considering the first case, and furthermore taking into account only lowest-order tunneling processes associated with sequential tunneling, the adiabatic pump works analogously to a peristaltic pump. The next-order correction turns out to be only due to a time-dependent renormalization of the dot-level position that is induced by the combination of Coulomb interaction and tunnel coupling to the leads. Remarkably this effect is not masked by other higher-order transport processes such as cotunneling. The situation is even more dramatic for pumping with the tunnel barriers. In this case, the lowest-order tunneling processes do not give rise to any pumping. The dominant pumping mechanism is, then, of higher order, namely pumping by making use of the time-dependent level renormalization. As a consequence, the gate-voltage dependence of the pumped charge provides a transparent experimental access to probe quantum-fluctuation effects.

The paper is organized as follows: Section II A introduces the model of the quantum dot. The time evolution of the system is described by a generalized Master equation in section II B and an adiabatic expansion is performed and applied to the current pumped through the dot in section II C. The expansion in the tunnel coupling is further discussed in section II D. The explicit evaluation of the formulae obtained up to here is done in section II E using a diagrammatic technique. In section III results for the pumped current and the pumped charge are presented and discussed.

II. MODEL AND FORMALISM

A. Model

We consider a single-level quantum dot with on-site Coulomb interaction coupled to two non-interacting leads. The system is described by the Hamiltonian

$$H = H_{\text{leads}} + H_{\text{dot}} + H_{\text{tun}}$$

where H_{leads} , H_{dot} , and H_{tun} describe, respectively, the left (L) and right (R) leads, the dot, and tunneling between dot and leads, and are given by

$$H_{\text{leads}} = \sum_{k,\sigma,\alpha} \epsilon_{k\alpha} c_{\sigma k\alpha}^\dagger c_{\sigma k\alpha} \quad (1a)$$

$$H_{\text{dot}} = \epsilon(t) \sum_{\sigma} n_{\sigma} + U n_{\uparrow} n_{\downarrow} \quad (1b)$$

$$H_{\text{tun}} = \sum_{k,\sigma,\alpha} \left[V_{\alpha}(t) c_{\sigma k\alpha}^\dagger d_{\sigma} + \text{h.c.} \right]. \quad (1c)$$

In Eqs. (1), $c_{\sigma k\alpha}$ ($c_{\sigma k\alpha}^\dagger$) is the fermionic annihilation (creation) operator for an electron with spin $\sigma = \uparrow, \downarrow$, momentum k , energy ϵ_k in lead $\alpha = \text{L, R}$; d_{σ} (d_{σ}^\dagger) is the fermionic annihilation (creation) operator for an electron with spin σ in the dot; and $n_{\sigma} = d_{\sigma}^\dagger d_{\sigma}$ is the number operator for the dot electrons with spin σ . The Coulomb interaction on the dot is described by the on-site energy U associated with double occupation. The leads are assumed to be in thermal equilibrium with the same chemical potential and to have flat bands with constant density of states ρ_{α} .

By periodically changing at least two of the system parameters, a DC current can be pumped through the dot. We choose the level position of the dot $\epsilon(t)$ and the tunnel matrix elements $V_{\alpha}(t)$ to be time-dependent. We only allow for the modulus, but not the phase, of $V_{\alpha}(t)$ to vary in time, since a time-dependent phase would correspond to a bias voltage. We define the time-dependent intrinsic line width $\Gamma_{\alpha}(t, t') = 2\pi \rho_{\alpha} V_{\alpha}(t) V_{\alpha}^*(t')$, the total intrinsic line width $\Gamma(t, t') = \Gamma_{\text{L}}(t, t') + \Gamma_{\text{R}}(t, t')$, as well as $\Gamma_{\alpha}(t) = \Gamma_{\alpha}(t, t)$ and $\Gamma(t) = \Gamma(t, t)$. To keep all formulae transparent, we set $\hbar \equiv 1$ throughout the paper.

B. Generalized Master equation and adiabatic approximation

As described above we consider an interacting quantum dot coupled to non-interacting leads. Since the leads act as baths, it is convenient to trace out the degrees of freedom of the non-interacting lead states to arrive at an effective description of the reduced system. In the limit of temperature much lower than the level spacing of the dot, only one level will contribute to transport. Therefore the Hilbert space for the dot is four dimensional: the quantum dot can be empty, singly occupied with a spin-up or a spin-down electron or doubly occupied. These states, labeled by $\chi = \{0, \uparrow, \downarrow, \text{d}\}$, have energy $E_0 = 0$, $E_{\uparrow} = E_{\downarrow} = \epsilon$ and $E_{\text{d}} = 2\epsilon + U$, respectively. In the following we use a matrix notation in the four dimensional Hilbert space of the dot, with boldface symbols for vectors and matrices. The probabilities to find the dot in the respective state are $\mathbf{p} = (p_0, p_{\uparrow}, p_{\downarrow}, p_{\text{d}})^T$.

The starting point of our analysis is the generalized Master equation for the time evolution of the system,

$$\frac{d}{dt} \mathbf{p}(t) = \int_{-\infty}^t dt' \mathbf{W}(t, t') \mathbf{p}(t'), \quad (2)$$

where the matrix elements $W_{\chi, \chi'}(t, t')$ of the kernel $\mathbf{W}(t, t')$ describe the transition from a state χ' at time t' to a state χ at time t . For the system considered here, Eq. (2) defines the most general kinetic equation for the dot probabilities without any approximation. Off-diagonal matrix elements of the reduced density matrix for the quantum dot, that correspond to *real* superposition of different states $\chi \neq \chi'$, do not couple to the diagonal ones since χ and χ' differ by a conserved quantum

number, particle number or spin. Therefore, off-diagonal elements of the reduced density matrix do not enter any transport quantity. Nevertheless, quantum fluctuation effects involving *virtual* intermediate states of the quantum dot in higher-order processes such as cotunneling, are fully taken into account by Eq. (2) by properly evaluating the kernel \mathbf{W} .³⁴

Our goal is to describe the response of the system to slow periodic variations of the system parameters $X(t)$ with frequency Ω . After waiting long enough, such that any memory of the initial dot-state distribution $\mathbf{p}(-\infty)$ has died out, the dynamics of the system is fully determined by the explicit time dependence of the system parameters. The latter enters Eq. (2) in two ways, namely by the kernel $\mathbf{W}(t, t')$ being a functional of the system parameters $X(\tau)$ with $\tau \in [t', t]$, and by the non-Markovian structure, i.e., the time derivative of $\mathbf{p}(t)$ at time t depends on $\mathbf{p}(t')$ at an earlier time t' at which the system parameters had different values. In the adiabatic regime it is possible to simplify considerably the form of the Master equation by performing an *adiabatic expansion*, i.e., an expansion in the pumping frequency Ω , assuming that Ω is small as compared to both the energy scales that determine the decay time of the kernel and the time integral of the kernel (which sets the time scale of the system's response to the parameter's change). The zeroth-order, *instantaneous*, term of the adiabatic expansion corresponds to freezing the value of all system parameters, $X(t)$, at time t , which corresponds to solving a time-independent problem. To obtain the first-order correction we need to systematically collect all contributions linear in the pumping frequency or, equivalently, linear in the time derivative of X at time t . For this, we perform a Taylor expansion of $\mathbf{p}(t')$ around t up to linear order,

$$\frac{d}{dt}\mathbf{p}(t) = \int_{-\infty}^t dt' \mathbf{W}(t, t') \left[\mathbf{p}(t) + (t' - t) \frac{d}{dt}\mathbf{p}(t) \right]. \quad (3)$$

Furthermore, we perform an adiabatic expansion of the kernel $\mathbf{W}(t, t')$ itself. The zeroth-order term, $\mathbf{W}_t^{(i)}(t - t')$, is indicated with the superscript (i) for *instantaneous* and the subscript t to emphasize that the system parameters $X(\tau) \rightarrow X(t)$ are frozen at time t . It depends only on the time difference $t - t'$, and only parametrically on t through $X(t)$. The first-order term is obtained by linearizing the time dependence of all parameters $X(\tau)$ with respect to the final time t , i.e., $X(\tau) \rightarrow X(t) + (\tau - t) \frac{d}{d\tau} X(\tau)|_{\tau=t}$, and retaining only linear terms in time derivatives. This linear correction to the kernel is indicated by the superscript (a) for *adiabatic*,

$$\mathbf{W}(t, t') \rightarrow \mathbf{W}_t^{(i)}(t - t') + \mathbf{W}_t^{(a)}(t - t'). \quad (4)$$

Finally, we need to perform an adiabatic expansion for the occupation probabilities in the dot,

$$\mathbf{p}(t) \rightarrow \mathbf{p}_t^{(i)} + \mathbf{p}_t^{(a)}. \quad (5)$$

The instantaneous probabilities $\mathbf{p}_t^{(i)}$ are the solution of the time-independent problem with all parameter values fixed at time t . They are obtained from the Master Eq. (3) in the stationary limit,

$$0 = \mathbf{W}_t^{(i)} \mathbf{p}_t^{(i)}, \quad (6)$$

together with the normalization condition $\mathbf{e}^T \mathbf{p}_t^{(i)} = 1$, where $\mathbf{e} = (1, 1, 1, 1)^T$, and we have introduced the Laplace transform

$$F(z) = \int_{-\infty}^t dt' e^{-z(t-t')} F(t - t')$$

to define $\mathbf{W}_t^{(i)} = \mathbf{W}_t^{(i)}(z = 0_+)$. The first adiabatic correction can be obtained from Eq. (3), using Eqs. (4) and (5). We find

$$\mathbf{W}_t^{(i)} \mathbf{p}_t^{(a)} = \frac{d}{dt} \mathbf{p}_t^{(i)} - \mathbf{W}_t^{(a)} \mathbf{p}_t^{(i)} - \partial \mathbf{W}_t^{(i)} \frac{d\mathbf{p}_t^{(i)}}{dt}, \quad (7)$$

where again $\mathbf{W}_t^{(i/a)}$ is the Laplace transform at zero frequency and $\partial \mathbf{W}_t^{(i)} = (\partial/\partial z) \mathbf{W}_t^{(i)}(z)|_{z=0_+}$. Once $\mathbf{W}_t^{(i/a)}$ are evaluated and the instantaneous probabilities $\mathbf{p}_t^{(i)}$ are known from Eq. (6), the adiabatic corrections $\mathbf{p}_t^{(a)}$ are obtained from Eq. (7) together with the normalization condition $\mathbf{e}^T \mathbf{p}_t^{(a)} = 0$.

C. Pumped charge

The charge Q pumped in one cycle $\mathcal{T} = 1/\Omega$ is related to the time-dependent current $I_L(t)$ flowing through the left barrier by

$$Q = \int_0^{\mathcal{T}} I_L(t) dt.$$

By accounting for the time evolution of the system before time t in a similar way as done for the dot-state probabilities above, we can express the current into the left lead as

$$I_L(t) = e \int_{-\infty}^t dt' \mathbf{e}^T \mathbf{W}^L(t, t') \mathbf{p}(t'), \quad (8)$$

where $\mathbf{W}^L(t, t') = \sum_p p \mathbf{W}^{Lp}(t, t')$, and $\mathbf{W}^{\alpha p}(t, t')$ includes all processes associated with transitions where the number of electrons (with charge e) entering reservoir α minus the ones leaving it equals p .

It is straightforward to perform an adiabatic expansion for Eq. (8) in the same way as for the Master equation. The instantaneous or zeroth-order level of the adiabatic expansion is sufficient to describe the DC current that is driven through the quantum dot by an applied transport voltage.³³ It is, furthermore, sufficient for modeling *rectification*, i.e., the generation of a DC current component by applying an AC transport voltage and appropriately

changing some system parameter in time. In the absence of any DC or AC transport voltage, as considered in the present paper, the instantaneous part of the current vanishes. In order to describe *pumping*, one needs to compute the first-order adiabatic correction of the current. Using for Eq. (8) the same procedure as for the Master equation, we find the adiabatic part of the current to be

$$I_L(t) = e \mathbf{e}^T \left[\mathbf{W}_t^{L(a)} \mathbf{p}_t^{(i)} + \mathbf{W}_t^{L(i)} \mathbf{p}_t^{(a)} + \partial \mathbf{W}_t^{L(i)} \frac{d\mathbf{p}_t^{(i)}}{dt} \right]. \quad (9)$$

D. Perturbation expansion in tunneling

Alongside with the adiabatic expansion we perform a perturbation expansion in powers of the tunnel coupling strength Γ for both the instantaneous and the adiabatic correction of the kernel \mathbf{W} , the probabilities \mathbf{p} , and the current $I_L(t)$. We indicate the order of the perturbation expansion in Γ by adding a superscript, i.e., $\mathbf{W}_t^{(i)} = \mathbf{W}_t^{(i,1)} + \mathbf{W}_t^{(i,2)} + \mathcal{O}(\Gamma^3)$ for the instantaneous contribution to the kernel, and similarly for $\mathbf{W}_t^{(a)}$. The expansion of the instantaneous probabilities begins in zeroth order in Γ , $\mathbf{p}_t^{(i)} = \mathbf{p}_t^{(i,0)} + \mathbf{p}_t^{(i,1)} + \mathcal{O}(\Gamma^2)$, in order to be able to fulfill the normalization condition $\mathbf{e}^T \mathbf{p}_t^{(i)} = 1$. By expanding Eq. (6) in powers of Γ , we find that the instantaneous probabilities should fulfill the two equations,

$$0 = \mathbf{W}_t^{(i,1)} \mathbf{p}_t^{(i,0)} \quad (10a)$$

$$0 = \mathbf{W}_t^{(i,2)} \mathbf{p}_t^{(i,0)} + \mathbf{W}_t^{(i,1)} \mathbf{p}_t^{(i,1)}, \quad (10b)$$

together with the normalization conditions $\mathbf{e}^T \mathbf{p}_t^{(i,0)} = 1$ and $\mathbf{e}^T \mathbf{p}_t^{(i,1)} = 0$. As discussed above, the instantaneous part of the current vanishes in all order in Γ due to the absence of an applied transport voltage.

In order to determine the adiabatic corrections to the probabilities, we expand also Eq. (7) in powers of Γ ,

$$\frac{d\mathbf{p}_t^{(i,0)}}{dt} = \mathbf{W}_t^{(i,1)} \mathbf{p}_t^{(a,-1)} \quad (11a)$$

$$\begin{aligned} \frac{d\mathbf{p}_t^{(i,1)}}{dt} = & \mathbf{W}_t^{(i,1)} \mathbf{p}_t^{(a,0)} + \mathbf{W}_t^{(i,2)} \mathbf{p}_t^{(a,-1)} \\ & + \mathbf{W}_t^{(a,1)} \mathbf{p}_t^{(i,0)} + \partial \mathbf{W}_t^{(i,1)} \frac{d\mathbf{p}_t^{(i,0)}}{dt}, \end{aligned} \quad (11b)$$

with the normalization conditions $\mathbf{e}^T \mathbf{p}_t^{(a,-1)} = 0$ and $\mathbf{e}^T \mathbf{p}_t^{(a,0)} = 0$. We emphasize that, in order to properly match the powers of Γ on the left and right hand side of Eq. (11), one has to start the expansion of the adiabatic correction of the probabilities in *minus first* order in Γ , $\mathbf{p}_t^{(a)} = \mathbf{p}_t^{(a,-1)} + \mathbf{p}_t^{(a,0)} + \mathcal{O}(\Gamma)$. At first glance, such an expansion might look divergent for the weak-coupling limit, $\Gamma \rightarrow 0$. However, in the validity range of the adiabatic expansion everything remains well defined: the

adiabaticity condition requires that the energy scale defined by the pumping frequency Ω is much smaller than the tunnel-coupling strength Γ . Since the time derivative on the left hand side of Eq. (11) introduces a factor Ω , we see that $\mathbf{p}_t^{(a,-1)}$ scales with Ω/Γ , which is always much smaller than 1 in the adiabatic limit.

The perturbation expansion of the adiabatically pumped current is derived from Eq. (9) and reads

$$I_L^{(0)}(t) = e \mathbf{e}^T \mathbf{W}_t^{L(i,1)} \mathbf{p}_t^{(a,-1)} \quad (12a)$$

$$\begin{aligned} I_L^{(1)}(t) = & e \mathbf{e}^T \left[\mathbf{W}_t^{L(i,1)} \mathbf{p}_t^{(a,0)} + \mathbf{W}_t^{L(i,2)} \mathbf{p}_t^{(a,-1)} \right. \\ & \left. + \mathbf{W}_t^{L(a,1)} \mathbf{p}_t^{(i,0)} + \partial \mathbf{W}_t^{L(i,1)} \frac{d\mathbf{p}_t^{(i,0)}}{dt} \right] \end{aligned} \quad (12b)$$

We see that the lowest-order contribution to the pumped current starts in zeroth order in the tunnel coupling strength Γ , as it consists in a product of a first- and a minus-first order term in the tunneling coupling, but scales linearly with the pumping frequency Ω . This contrasts with the DC current driven by a finite bias voltage, for which the lowest-order contribution is linear in Γ .

Certain properties of pumping can be derived by a closer inspection of the perturbative expansion of the Master equation. To zeroth order in Γ the pumped current is nonzero only if $\mathbf{p}^{(a,-1)}$ is non vanishing, which, according to Eq. (11a) requires that the zeroth-order instantaneous probabilities $\mathbf{p}^{(i,0)}$ depends on time. However the latter are simply determined by the Boltzmann factors of the corresponding state energies:

$$p_\chi^{(i,0)} = \frac{e^{-\beta E_\chi}}{Z},$$

where E_χ is the energy related to the dot state χ , $\beta = 1/k_B T$ is the inverse temperature and Z the partition function. In particular, the probabilities $p_\chi^{(i,0)}$ are independent of the tunnel couplings. As a consequence, in order to have a non-vanishing zeroth-order pumped current $I_L^{(0)}$, one of the pumping parameters has to be the level position. When pumping with the two barrier heights, $I_L^{(0)}$ vanishes.

E. Diagrammatic rules

In order to evaluate the kernel \mathbf{W} of the Master equation Eq. (2) we use the diagrammatic perturbation approach to transport through interacting quantum dots developed in Ref. 33. While in Ref. 33 the diagrammatic language was derived for DC transport with time-independent system parameters, we generalize the approach in this section to account for the adiabatic time dependence of the external parameters.

We start with deriving the Master equation from a very general point of view in order to relate its kernel to a set of diagrams to be evaluated. In general, the (time-dependent) transport properties are governed by the time

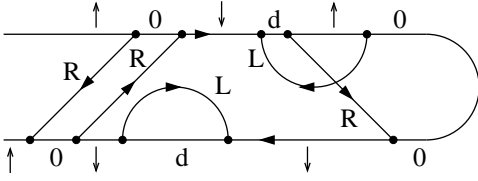


FIG. 1: Example for the time evolution of the reduced density matrix. The upper and lower line represent the forward and backward time propagation along the Keldysh contour. Tunneling lines connecting vertices represent tunneling events with the left (right) reservoir. Next to the propagators the respective dot states are indicated.

evolution of the reduced density matrix of the dot obtained after tracing out the degrees of freedom of the non-interacting lead electrons. Since the leads are non-interacting and in thermal equilibrium, the lead electrons can be integrated out making use of Wick's theorem, i.e. contracting pairs of creation and annihilation operators $c_{\sigma k \alpha}^\dagger (c_{\sigma k \alpha})$. Furthermore, since in the case of pumping there is no voltage applied between left and right lead, the occupation of electronic states in both leads is described by the same Fermi distribution function $f(\omega)$. The time evolution of the reduced density matrix is related to the propagator $\Pi(t, t')$ by

$$\mathbf{p}(t) = \Pi(t, t') \mathbf{p}(t'). \quad (13)$$

Contributions to this propagator can be depicted as diagrams on the Keldysh contour, where contractions of fermion operators of the leads are indicated as tunneling lines. An example is shown in Fig. 1. The propagator $\Pi(t, t')$ can be expressed in terms of its irreducible part $\mathbf{W}(t''', t'')$ by means of a Dyson equation:

$$\Pi(t, t') = \mathbf{1} + \int_{t'}^t dt''' \int_{t'}^{t'''} dt'' \mathbf{W}(t''', t'') \Pi(t'', t'). \quad (14)$$

The irreducible diagram part $\mathbf{W}(t''', t'')$ is defined as the sum over all diagrams in which any vertical cut crosses at least one tunneling line (see Fig. 2 as an example).

Performing the time derivative of Eq. (13), plugging in Eq. (14), and shifting the lower bound of the remaining time integral to minus infinity, we obtain the generalized Master equation of Eq. (2).

In a similar way we proceed for the current $I_\alpha(t) = e \frac{d}{dt} \langle N_\alpha(t) \rangle$ of particles flowing into reservoir α , which is given by

$$I_\alpha(t) = -ie \sum_{k, \sigma} \left[V_\alpha \langle c_{\sigma k \alpha}^\dagger d_\sigma \rangle(t) - V_\alpha^* \langle d_\sigma^\dagger c_{\sigma k \alpha} \rangle(t) \right]. \quad (15)$$

It can also be expressed in terms of diagrams. They contain a vertex at time t that has the same structure as the tunneling vertices in Fig. 1, and can be attached to the upper or the lower propagator line. We find Eq. (8) where $\mathbf{W}^\alpha(t, t') = \sum_p p \mathbf{W}^{\alpha p}(t, t')$ has the following properties.

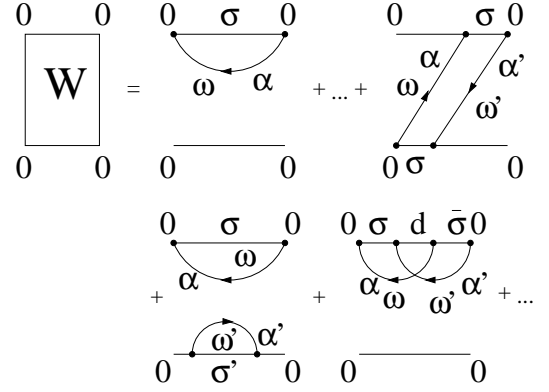


FIG. 2: Examples of diagrams contributing to the irreducible diagram part W_{00} .

The terms $\mathbf{W}^{\alpha p}(t, t')$ are given by all diagrams for which the number of tunneling lines with reservoir index α running from the upper to the lower propagator minus the number of those with reservoir index α running in the opposite direction equals p .

In the following we summarize the diagrammatic rules for the kernel $\mathbf{W}_t^{(i)}$ (as described in Ref. 33) and discuss additional rules for the evaluation of its adiabatic expansion. Examples for the application of the rules given below are shown in appendix A. We start with rules for the Laplace transform of $\mathbf{W}_t^{(i, n)}(z) = \int_{-\infty}^t dt' \exp(-z(t-t')) \mathbf{W}_t^{(i, n)}(t, t')$ in n -th order in the tunnel coupling. This will directly lead us to the desired objects $\mathbf{W}_t^{(i, n)} = \mathbf{W}_t^{(i, n)}(z)|_{z=0+}$ and $\partial \mathbf{W}_t^{(i, n)} = (\partial/\partial z) \mathbf{W}_t^{(i, n)}(z)|_{z=0+}$.

(1) Draw all topologically different diagrams with n directed tunneling lines connecting pairs of vertices containing lead electron operators. Assign a reservoir index α , an energy ω and a spin index σ to each of these lines. Assign states χ and the corresponding energies $E_\chi(t)$ to each element of the Keldysh contour connecting two vertices. Furthermore, draw an external line from the upper leftmost beginning of a dot propagator to the upper rightmost end of a dot propagator that carries the (imaginary) energy $-iz$.

(2) For each time segment between two adjacent vertices (independent on whether they are on the same or on opposite branches of the Keldysh contour) write a resolvent $1/\Delta E(t)$ where $\Delta E(t)$ is the difference of left going minus right going energies (including energies of tunneling lines and the external line - the positive imaginary part of iz will keep all resolvents regularized).

(3) Each vertex containing a dot operator $d_\sigma^{(\dagger)}$ gives rise to a matrix element $\langle \chi' | d_\sigma^{(\dagger)} | \chi \rangle$ where χ (χ') is the dot state entering (leaving) the vertex with respect to the Keldysh contour.

(4) The contribution of a tunneling line of reservoir α is $\frac{1}{2\pi} \Gamma_\alpha(t) f(\omega)$ if the line is going backward with respect to the closed time path and $\frac{1}{2\pi} \Gamma_\alpha(t) [1 - f(\omega)]$ if it is going forward.

- (5) The overall prefactor is given by $(-i)(-1)^{b+c}$ where b is the total number of vertices on the backward propagator and c the number of crossings of tunneling lines.
- (6) Integrate over the energies of tunneling lines and sum over reservoir and spin indices.

To derive the rules for the adiabatic corrections $\mathbf{W}_t^{(a,n)}$ we first analyze how the time-dependent parameters enter the expression of the kernel $\mathbf{W}(t, t')$. The time-dependent variables for which the adiabatic expansion has to be performed are $V_\alpha(t)$ and $\epsilon(t)$, where the first one only appears in the product $\Gamma_\alpha(t_i, t_j) = 2\pi\rho V_\alpha(t_i) V_\alpha(t_j)$ associated with a tunneling line, and the latter only in the isolated-dot propagator $\exp\left(-i \int_{t_i}^{t_j} dt' E_\chi(t')\right)$ for each segment between adjacent vertices. While for the instantaneous kernels all parameters were taken at time t , now we perform a series expansion around the same time t and keep all contributions linear in a time derivative of the pumping parameters,

$$\Gamma(t_i, t_j) \approx \Gamma(t) + \frac{t_i - t}{2} \frac{d\Gamma}{dt}(t) + \frac{t_j - t}{2} \frac{d\Gamma}{dt}(t) \quad (16)$$

$$e^{-i \int_{t_i}^{t_j} dt' E_\chi(t')} \approx e^{-i E_\chi(t) \cdot (t_j - t_i)} \times \left[1 - i \frac{(t_j - t)^2 - (t_i - t)^2}{2} \frac{dE_\chi}{dt}(t) \right] \quad (17)$$

The factors $(t_i - t)$ or $(t_j - t)^2$ can be included in the diagrammatic rules in the following way: introduce an additional external frequency line with the imaginary energy $-iz_i$ from the vertex at t_i to the rightmost vertex at t (or the imaginary energy $-iz_j$ from the beginning of a dot propagator line at t_j to the rightmost upper end of a dot propagator line at t), performing the first derivative with respect to z_i (or second derivative with respect to z_j) then set $z_i = 0_+$ and $z_j = 0_+$. The external frequency lines are drawn as dotted lines in Fig. 3.

The rules to compute the contribution to the adiabatic corrections $\mathbf{W}_t^{(a,n)}$ due to the time-dependence of $\Gamma(t)$ read:

- (7a) Add to all diagrams needed for $\mathbf{W}_t^{(i,n)}(z)$ additional external frequency lines between any vertex t_i and the right corner of the diagram and assign to them an (imaginary) energy $-iz_i$. Note that an eventual external frequency line between two right corners of a diagram does not contribute and can always be omitted.
- (7b) Follow the rules (1) to (6) taking into account the extra lines.
- (7c) Perform a first derivative with respect to z_i and multiply it by the factor $\frac{1}{2} \frac{d\Gamma}{dt}(t) \frac{1}{\Gamma(t)}$. Sum all the contributions obtained in this way.
- (7d) Set all the external frequencies z_i and z to 0_+ .

The contribution to the adiabatic correction $\mathbf{W}_t^{(a,n)}$ due to the time-dependence of the level position can be

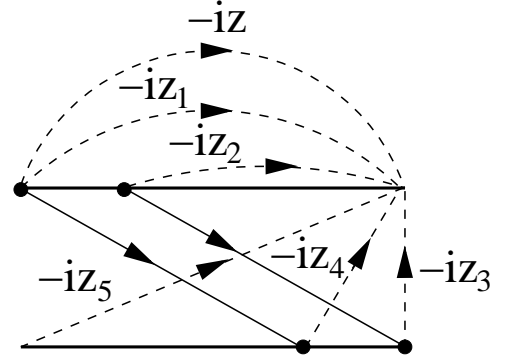


FIG. 3: Example on how to add the external frequency lines. The line $-iz$ is needed for the evaluation of $\partial \mathbf{W}$; $-iz_1$, $-iz_2$, and $-iz_4$ are needed for the contributions to $\mathbf{W}^{(a)}$ due to both $\frac{d\Gamma}{dt}$ and $\frac{d\epsilon}{dt}$; $-iz_3$ does not contribute and can be omitted; and $-iz_5$ is additionally needed for the evaluation of the contribution to $\mathbf{W}^{(a)}$ due to $\frac{d\epsilon}{dt}$.

computed in a similar way:

(8a) In addition to the external frequency lines added according to rule (7a), put one more external frequency line from the left corner of the diagram with no vertex to the right corner.

(8b) Follow the rules (1) to (6) taking into account the extra lines.

(8c) Perform a second derivative with respect to z_i and multiply by $-\frac{i}{2} \frac{d(E_\chi - E_{\chi'})}{dt}(t)$, where χ (χ') is the dot state entering (leaving) the vertex of the external frequency line at t_i with respect to the Keldysh contour. The term $\frac{dE_\chi}{dt} \left(\frac{dE_{\chi'}}{dt} \right)$ is omitted if the segment associated with E_χ ($E_{\chi'}$) does not belong to the diagram. Sum all the contributions obtained in this way.

(8d) Set the external frequencies z_i and z to 0_+ .

III. RESULTS

In this section we show the results for the pumped current and the pumped charge through a single-level quantum dot. As no bias voltage is applied, the only contribution to the pumped current arises from the adiabatic correction, and hence we drop the superscript (a) for the current. As we discuss below, the properties of the pumped charge, in the regime discussed in this paper, can be understood to a large extent in terms of the time-dependence of the occupation of the quantum dot, $\langle n \rangle = p_\uparrow + p_\downarrow + 2p_d$. For this reason, we first want to discuss the perturbation expansion of the instantaneous average charge occupation. The contribution to zeroth order in Γ turns out to be determined by the Boltzmann factors of the energies associated with the states of the isolated quantum dot. This yields

$$\langle n \rangle^{(i,0)} = \frac{2f(\epsilon)}{1 + f(\epsilon) - f(\epsilon + U)}. \quad (18)$$

The first-order correction accounts for quantum fluctuations due to tunneling from and to the leads. There are two qualitatively different effects which are due to tunneling and correspondingly we present the results for the first-order corrections as a sum of two contributions $\langle n \rangle^{(i,1)} = \langle n \rangle^{(i,broad)} + \langle n \rangle^{(i,ren)}$, where the contribution to broadening is the sum of the contributions of the two leads $\langle n \rangle^{(i,broad)} = \langle n \rangle^{(i,broad,R)} + \langle n \rangle^{(i,broad,L)}$.

First, the dot levels acquire a finite life-time broadening due to the coupling to lead α , which is accounted for by

$$\langle n \rangle^{(i,broad)} = \left(2 - \langle n \rangle^{(i,0)}\right) \phi'(\epsilon) + \langle n \rangle^{(i,0)} \phi'(\epsilon + U), \quad (19)$$

where $\phi'(\omega)$ is the derivative of $\phi(\omega) = \frac{\Gamma}{2\pi} \text{Re} \Psi\left(\frac{1}{2} + \frac{i\beta\omega}{2\pi}\right)$, and Ψ is the digamma function. The first term accounts for the broadening of the resonance at ϵ between empty and singly-occupied dot; it has a prefactor 2, when the dot is empty, and is zero when the dot is doubly occupied. The second term accounts for the broadening of the resonance at $\epsilon + U$ between singly- and doubly-occupied dot (this contribution is zero if the dot is empty). In the case that the dot is singly occupied both terms contribute with a prefactor 1.

Second, the combination of tunneling and charging energy gives rise to a renormalization of the level position, $\epsilon \rightarrow \epsilon + \sigma(\epsilon, \Gamma, U)$ with

$$\sigma(\epsilon, \Gamma, U) = \phi(\epsilon + U) - \phi(\epsilon), \quad (20)$$

as it is expected from the poor man's scaling analysis.³⁵ The level renormalization is positive when the level is in the vicinity of the Fermi energy of the leads and negative for $\epsilon + U$ being close to the Fermi energy. This means that the distance between the two resonances from empty to singly-occupied dot and from singly- to doubly-occupied dot is effectively decreased. The changes to the instantaneous average occupation due to the level renormalization reads

$$\langle n \rangle^{(i,ren)} = \frac{d}{d\epsilon} \left(\langle n \rangle^{(i,0)} \right) \sigma(\epsilon, \Gamma, U). \quad (21)$$

The sum of Eqs. (19) and (21) is directly found from the correction in first order Γ to the occupation probability, which is shown explicitly in appendix B. We remark that the above interpretation of the two terms is in full agreement with known exact results for the non-interacting case. For $U = 0$ (and flat density of states in the leads), the level renormalization vanishes. The spectral density is then equal to the Breit-Wigner function and its expansion in zeroth and first order in Γ leads to the non-interacting result of $\langle n \rangle^{(i,0)}$ and $\langle n \rangle^{(i,broad)}$.

A. Adiabatically-pumped current

We now proceed with solving Eqs. (10a), (11a) and (12a) for arbitrary interaction U to get the zeroth-order

adiabatically pumped current. The result of the diagrammatic approach explained above can be written in the form

$$I_L^{(0)}(t) = -e \frac{\Gamma_L}{\Gamma} \frac{d}{dt} \langle n \rangle^{(i,0)}. \quad (22)$$

This suggests the following interpretation. As the dot occupation is changed in time by varying the pumping parameters (one of them must be the level position since $\langle n \rangle^{(i,0)}$ is independent of the tunnel-coupling strengths), the charge moves in and out of the quantum dot generating a current from/into the leads. The contributions flowing through barrier α split weighted by the time-dependent relative tunnel couplings Γ_α/Γ .

By means of Eqs. (10b), (11b) and (12b), one finds the first-order- Γ contribution to the current,

$$I_L^{(1)}(t) = -e \left\{ \frac{d}{dt} \left(\langle n \rangle^{(i,broad,L)} \right) + \frac{\Gamma_L}{\Gamma} \frac{d}{dt} \langle n \rangle^{(i,ren)} \right\}. \quad (23)$$

Again, we have written the result in such a form that an identification of the pumping mechanism is straightforward. The first term of Eq. (23) contains the contribution due to the correction of the average dot occupation induced by the tunnel coupling to the left lead. Intuitively the finite-life-time broadening due to the coupling to the left lead is associated with tunnel processes of electrons through the left barrier. Any change in the life-time broadening due to coupling to the left lead will, therefore, result in a current through the left barrier only. As a result, this first term contains a total time derivative, and as parameters are periodically changing in time, it will not lead to a net pumped charge after the full pumping cycle. We conclude that changing the life-time broadening of the dot level does not contribute to adiabatic pumping. The second term has the same structure as the zeroth-order contribution, Eq. (23). It can be understood as the correction term introduced by renormalizing the position of the dot level, which may be time dependent via time-dependent tunnel couplings or a time-dependent gate voltage. Again, the charge transferred in/out of the quantum dot splits into two currents to or from both leads with relative weight Γ_α/Γ . Now, even if the dot level is constant and only both the tunnel couplings Γ_α are varying in time, a finite charge can be pumped by means of level renormalization.

It is useful to compare these findings with a perturbation expansion of the DC current driven by a DC transport voltage. In lowest (first) order, current is carried by sequential-tunneling processes. A systematic calculation of the second-order linear conductance³⁶ shows that quantum fluctuation due to tunneling give rise to three different types of correction terms. The first one, which dominates the linear conductance in the Coulomb-blockade regime away from resonance, is due to cotunneling. One way to depict cotunneling is to understand it as transport through the finite-life-time broadened dot level. It would, thus, correspond to the first term of Eq. (23).

Close to resonance, however, there are two more corrections to the sequential-tunneling linear conductance. They can be cast as sequential tunneling but with renormalized level position, as discussed above, or with renormalized tunnel coupling strength. For the DC current, all these three contributions are present at the same time, which makes it challenging to identify them separately in an experiment. For the adiabatically pumped charge, where correction terms associated with a renormalization of the tunnel couplings and level-broadening effects vanish, the situation is distinctively different. Studying adiabatic pumping is, therefore, a convenient tool to access the energy-level renormalization. This is most dramatic in the case when the zeroth-order pumped current is zero, i.e., when pumping is done by changing both tunnel couplings. In this case, the *dominant* contribution to the pumped charge is due to time-dependent level renormalization.

B. Weak pumping

When writing the pumped charge, we report as indices, in the following, the particular choice of pumping parameters it refers to. For example if the pumping fields are Γ_L and ϵ , we indicate the charge as $Q_{\Gamma_L, \epsilon}$. We now concentrate on weak pumping. We write the time-dependent parameters in the form $\epsilon(t) = \bar{\epsilon} + \Delta\epsilon(t)$ and $\Gamma_\alpha(t) = \bar{\Gamma}_\alpha + \Delta\Gamma_\alpha(t)$ (with $\Delta\epsilon(t)$ and $\Delta\Gamma_\alpha(t)$ having zero time average) and expand the current up to bilinear response in the time-dependent part of the parameters. Choosing ϵ and Γ_L as pumping parameters we obtain up to first order in Γ :

$$Q_{\Gamma_L, \epsilon} = -e \frac{\bar{\Gamma}_R}{\bar{\Gamma}^2} \eta_1 \frac{d}{d\bar{\epsilon}} \left[\langle \bar{n} \rangle^{(i,0)} + \sigma(\bar{\epsilon}, \bar{\Gamma}, U) \frac{d}{d\bar{\epsilon}} \langle \bar{n} \rangle^{(i,0)} \right], \quad (24)$$

where the prefactor η_1 characterizes the amplitudes of the pumping parameters as well as their relative phase:

$$\eta_1 = \int_0^T \frac{\partial \Delta\epsilon}{\partial t} \Delta\Gamma_L dt.$$

It was already pointed out for a noninteracting system in Ref. 4 that in the limit of weak, adiabatic pumping the pumped charge is proportional to the surface enclosed in parameter space during one pumping cycle, which in this case is equal to η_1 . Furthermore, $\langle \bar{n} \rangle^{(i,0)}$ is the instantaneous occupation of the dot computed with the time-dependent parameters taken at their time-average value, and $\bar{\Gamma} = \bar{\Gamma}_L + \bar{\Gamma}_R$. The first term inside the brackets is the zeroth-order- Γ contribution to the charge and, therefore, it is the dominant one. It has two peaks as a function of the average level position, which are located, in the limit $U \gg k_B T$, at $\bar{\epsilon} = -U - k_B T \ln(2)$ and $\bar{\epsilon} = k_B T \ln(2)$. The second term is first order in Γ , stems from level renormalization, and vanishes in the non-interacting case. The first-order- Γ correction tends to decrease the distance between the two resonances.

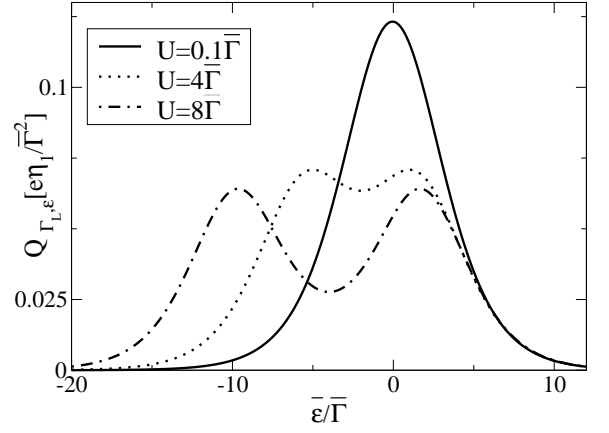


FIG. 4: Pumped charge up to first order Γ in units of $e\eta_1/\bar{\Gamma}^2$ as a function of the time-average level position in units of $\bar{\Gamma}$ for different values of U . Pumping parameters are ϵ and Γ_L . The temperature is $k_B T = 2\bar{\Gamma}$.

In Fig. 4 the pumped charge of Eq. (24) is shown in units of $e\eta_1/\bar{\Gamma}^2$ as a function of the time-average value of the level position, for different strengths of the interaction. The two peaks are directly related to transitions between singly-occupied and empty dot and between doubly- and singly-occupied dot. The shift by $k_B T \ln(2)$ of the peak positions is of combinatoric origin and reflects the fact that the probability of single occupation of the dot is increased due to the two spin states which lead to the same occupation number. Both peaks contribute with the same sign, as expected since the underlying pumping mechanism is the same for both resonances: in both cases the dot filling increases (decreases) when the level position is decreased (increased).

We now focus our attention on pumping with the two tunnel-coupling strengths Γ_L and Γ_R . In this case, there is no zeroth-order- Γ contribution to the pumped charge, as discussed above. The contribution to first order in Γ reads

$$Q_{\Gamma_L, \Gamma_R} = e \frac{\eta_2}{\bar{\Gamma}^2} \frac{d}{d\bar{\epsilon}} \left(\langle \bar{n} \rangle^{(i,0)} \right) \sigma(\bar{\epsilon}, \bar{\Gamma}, U), \quad (25)$$

where $\eta_2 = \int_0^T \frac{\partial \Delta\Gamma_L}{\partial t} \Delta\Gamma_R dt$ accounts for the pumping-parameter amplitudes and their relative phase as discussed in detail before for the quantity η_1 . The result for the pumped charge as a function of the level position is shown in Fig. 5. The solid line shows the result for very small interaction. As expected, it tends to zero, because the level renormalization vanishes. In the presence of interaction two peaks appear, which separate for increasing U . The two peaks are related to the two resonances at the level positions. They contribute with opposite sign. This reflects the opposite sign of the level renormalization for the two resonances. We remind that the first non-vanishing contribution of the perturbation expansion to the charge pumped through the dot by periodic change of the tunnel barriers is uniquely due to the effect of level renormalization. The height of the peaks increases for

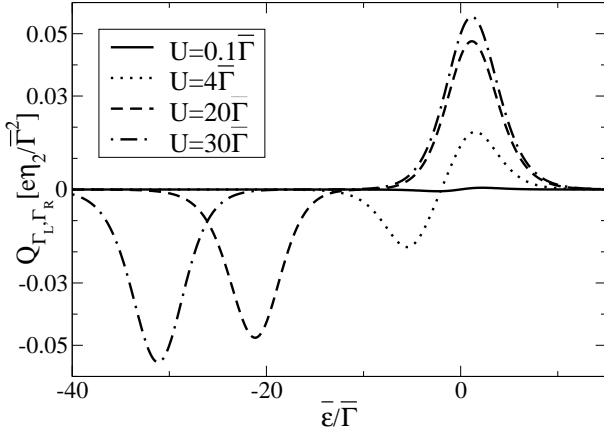


FIG. 5: Pumped charge up to first order Γ in units of $e\eta_2/\bar{\Gamma}^2$ as a function of the time-average level position in units of $\bar{\Gamma}$ for different values of U . Pumping parameters are Γ_L and Γ_R . The temperature is $k_B T = 2\bar{\Gamma}$.

increasing U , growing logarithmically for large U . Eventually, this increase will be cut-off by the bandwidths D , which we here chose to be infinite. The different sign of the pumped charge for the two resonances could serve as a signature to distinguish level-renormalization-induced pumping from parasitic peristaltic pumping due to cross capacitances of the gates modulating the tunnel couplings to the quantum dot.

IV. CONCLUSIONS

We have presented a perturbative approach in tunneling to adiabatic pumping through interacting quantum dots. In particular, a general diagrammatic technique to perform the adiabatic expansion has been developed. This technique has been applied to compute the pumped charge through a single-level quantum dot at temperatures much higher than the Kondo temperature. Two pumping schemes have been considered: pumping with the level position and one tunnel barrier, and pumping with the two barriers. When pumping with the level position and one tunnel barrier, the dominant mechanism of the adiabatic pump works analogously to a peristaltic pump. The next-order correction is related to the level renormalization induced by the interplay of Coulomb repulsion and electron tunneling. The situation is far more interesting for the case of pumping with the two barriers. With this pumping scheme there is no pumping in lowest order in the tunnel coupling, and the first non-vanishing contribution is due to the time-dependent level renormalization. Hence, we have demonstrated the importance of level-renormalization effects in pumping through interacting quantum dots. In particular, our results suggest that adiabatic pumping can be used to gain experimental access to the level renormalization in quantum dots.

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APPENDIX A: EXAMPLES OF DIAGRAMS

In this section we show how to apply the diagrammatic rules for the matrix element $(\mathbf{W}_t)_{0,0}$.

We start with the instantaneous term to lowest order in the tunnel coupling, $(\mathbf{W}_t^{(i,1)}(z))_{0,0}$. The corresponding diagrams are shown in Fig. 6. Two topologically different diagrams contribute, and each of them has to be summed over the spin index σ and the lead index α , and to be integrated over ω . We obtain:

$$(\mathbf{W}_t^{(i,1)}(z))_{0,0} = -i \sum_{\sigma,\alpha} \int \frac{d\omega}{2\pi} \left[\frac{\Gamma_\alpha f(\omega)}{\omega - \epsilon + iz} + \frac{\Gamma_\alpha f(\omega)}{\epsilon - \omega + iz} \right].$$

By letting $z = 0_+$ and making use of $1/(x + i0_+) = P/x - i\pi\delta(x)$, where P indicates Cauchy's principal value, we get

$$\begin{aligned} (\mathbf{W}_t^{(i,1)})_{0,0} &= -2\Gamma f(\epsilon), \\ (\partial \mathbf{W}_t^{(i,1)})_{0,0} &= -\frac{2\Gamma}{\pi} \frac{d}{d\epsilon} \int_P d\omega \frac{f(\omega)}{\omega - \epsilon}. \end{aligned}$$

For the adiabatic correction we need to introduce additional external frequency lines according to the rules 7a and 8a (see Fig.6). The additional line of rule 8a, going from the left corner of the diagram with no vertex to the right corner, does not contribute in this case and we have omitted drawing it. The evaluation of these diagrams leads us to the result

$$(\mathbf{W}_t^{(a,1)})_{0,0} = -\frac{d\Gamma}{dt} \frac{d}{d\epsilon} \int_P d\omega \frac{f(\omega)}{\omega - \epsilon} - \frac{\Gamma \frac{d\epsilon}{dt}}{\pi} \frac{d^2}{d\epsilon^2} \int_P d\omega \frac{f(\omega)}{\omega - \epsilon}$$

We now calculate the second-order- Γ contribution to the same matrix element of the instantaneous kernel. All diagrams contributing to the matrix element $(\mathbf{W}_t^{(i,2)})_{0,0}$ are depicted in Fig. 7. We sum over all appearing indices

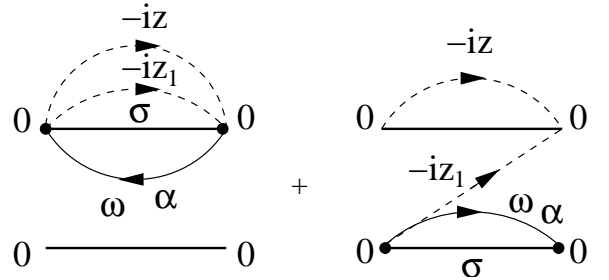


FIG. 6: Diagrams contributing to the first-order- Γ part of the adiabatic correction to the matrix element $(\mathbf{W}_t)_{0,0}$. All appearing reservoir and spin indices α, σ are to be summed over and the energy ω is to be integrated over.

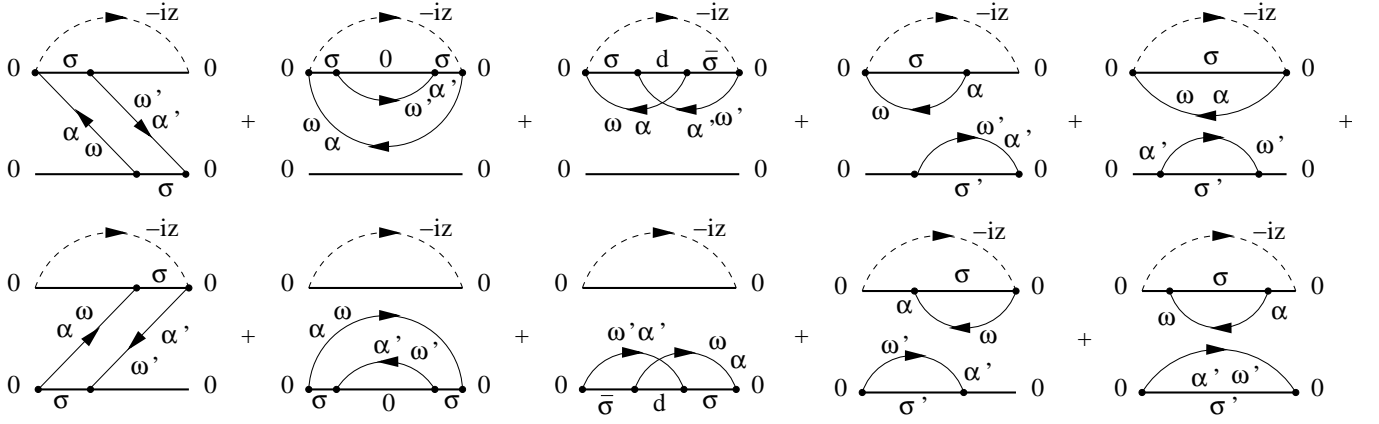


FIG. 7: Diagrams contributing to the second-order- Γ instantaneous part of the matrix element $(\mathbf{W}_t)_{0,0}$. All appearing reservoir and spin indices $\alpha, \alpha', \sigma, \sigma'$ are to be summed over and the energy ω is to be integrated over.

$\alpha, \alpha', \sigma, \sigma'$. The variables $\bar{\sigma}$ denotes the opposite spin of σ . As an example, we report the result for the sum of the first diagram of the first and the second line (after setting $z = 0_+$):

$$\frac{\Gamma^2}{\pi} \left[2f(\epsilon) \frac{d}{d\epsilon} \int_{\mathbb{P}} d\omega \frac{f(\omega)}{\omega - \epsilon} - \frac{d}{d\epsilon} \int_{\mathbb{P}} d\omega \frac{(f(\omega))^2}{\omega - \epsilon} \right].$$

To obtain the full second-order contribution $(\mathbf{W}_t^{(i,2)})_{0,0}$ we need to evaluate the remaining diagrams in Fig. 7 along the same lines as discussed in this Appendix.

APPENDIX B: OCCUPATION PROBABILITIES

Some intermediate results were not presented in the main part of this article as they were lengthy or not immediately necessary for the interpretation of the pumped current. Here we discuss in detail the first corrections to the occupation probabilities. The corrections to the occupation probabilities are used for the evaluation of Eq. (12) but do not appear directly in the results for the pumped current. We find for $\mathbf{p}_t^{(a,-1)}$, the first order adiabatic correction in minus first order in Γ :

$$\mathbf{p}_t^{(a,-1)} = -\frac{d\mathbf{p}_t^{(i,0)}}{dt} \frac{1}{2\Gamma} \frac{1}{[1 + f(\epsilon) - f(\epsilon + U)]}.$$

The adiabatic correction of the occupation probability is proportional to the time-derivative of $\mathbf{p}_t^{(i,0)}$, and is there-

fore an eigenvector of the matrix $\mathbf{W}_t^{(i,1)}$. The sign of this correction depends on the sign of the time derivative of $\epsilon(t)$.

The first-order- Γ correction to the instantaneous occupation probability is:

$$\begin{aligned} \mathbf{p}_t^{(i,1)} &= \frac{d\mathbf{p}_t^{(i,0)}}{d\epsilon} \sigma(\epsilon, \Gamma, U) \\ &+ \left(2 - \langle n \rangle^{(i,0)}\right) \phi'(\epsilon) \left(-1, \frac{1}{2}, \frac{1}{2}, 0\right)^T \\ &+ \langle n \rangle^{(i,0)} \phi'(\epsilon + U) \left(0, -\frac{1}{2}, -\frac{1}{2}, 1\right)^T. \end{aligned}$$

It consists of a part due to level renormalization (first row) and a part due to level broadening (second and third row). The correction due to level renormalization affects all four probabilities in the same functional way.

The correction due to broadening has two contributions. The first one is related to the broadening due to fluctuations between empty and singly-occupied dot. It is zero in the case that the dot is doubly occupied and largest when the dot is empty. The second contribution is related to the broadening due to fluctuations between singly- and doubly-occupied dot. It is zero when the dot is empty and largest when the dot is doubly occupied.

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